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MICROSTRUCTURE REACTOR
[Mikrostruktur-Reaktor]

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This invention relates to a microstructure reactor for carrying out endothermic chemical reactions in the presence of a catalyst (1) that is electrically heatable by resistance heating, whereby the reaction chamber of the reactor comprises a plurality of reaction channels (2) that are worked into a silicon substrate (3) and have cross-sectional dimensions in the submillimeter range. Catalyst (1) is applied at a uniform layer thickness on the inside surface of reaction channels (2).

Description

This invention relates to a microstructure reactor for carrying out endothermic chemical reactions in accordance with the precharacterizing part of Claim 1.

Reactors for endothermic processes in the chemical industry generally have relatively large dimensions and, as a result, a large mass, which require expensive heating for carrying out the desired reaction. The result is that, dynamically speaking, such reactors are very lethargic and temperature changes can occur only very slowly. Another disadvantage is that it takes a significant amount of time to turn such reactors on and off.

An additional problems with conventional reactors in which heat must be made available for an endothermic reaction is the poor heat transfer from a heating medium, through the reactor wall, into the process medium. When the heat must be introduced into the reaction process from the outside, heat losses commensurate with the quality of the heat transfer must be expected. In many cases, this problem can be alleviated if the heat for the endothermic reaction is provided

directly in the reactor by a controlled exothermic reaction. This can be achieved by selective partial combustion of the process medium. In this case, there is no need for heat transfer through the reactor wall. In this regard, the device is operated more dynamically. The disadvantage of this is that the waste gases from combustion mix with the chemical products formed in the endothermic reaction. Consequently, costly separation and purification processes are required. It should also be noted that the endothermic reaction occurring in the reactor can be influenced by the combustion.

Proposals have been made for remedying these problems, the primary goal of which is to drastically reduce the mass of the reactor, thereby significantly improving their dynamics. Such approaches to a solution are found in the development of so-called microreactors or microstructure reactors. From the publication "Entwicklung von Mikrostruktur-Apparaten für Anwendungen in der chemischen und thermischen Verfahrenstechnik" [Development of Microstructure Devices for Application in Chemical and Thermal Process Engineering] (Wissenschaftliche Berichte [Scientific Reports] FZKA 6080, Forschungszentrum Karlsruhe [Research Center Karlsruhe], 1998) microstructure reactors are known that are made of metallic materials such as copper, aluminum, silver, or stainless steel. Such units, which are made, for example, as cross-flow-type heat exchangers, have a plurality of passage channels for the process media, whose cross-sectional dimensions lie in the submillimeter range (e.g., 70x100 μm).

Using a cubic volume of only 3 cm edge length, a heat-exchange power in the range of 200 kW has been achieved with a process medium of water in pure heat-exchange operation, with a throughput of 7,000 kg/h. The present publication will report that, in principle, these apparatuses are also suitable for carrying out chemical reactions.

In some research (e.g., at the University of Stuttgart), microstructure reactors have been built in which silicon has been used instead of metallic materials. As in the construction of electronic components of silicon, it is relatively easy to use etching to produce very small channels that can be used for passing a process medium in the manner of a reaction chamber. This publication will report on a corresponding microreactor chamber having a cross-sectional area of 500x500 µm and a length of ca. 20 mm. In order to utilize such a reactor for an endothermic catalytic reaction, each reaction channel is to be provided with a precious-metal wire 100 µm thick, extending coaxially through the reaction channel and operating as a catalyst. The heat required for the endothermic reaction is not brought into the microstructure reactor by a heat-exchange medium, but is given off by electrical-resistance heating from the inserted precious-metal wire. The disadvantage of this solution is that the catalytically active thin precious-metal wire provides only a relatively small surface area. Moreover, the position of the precious-metal wire in the reaction channel, which is coaxial in the optimum case, can be

significantly altered by intense thermal expansion, resulting in disadvantages for the desired chemical reaction.

The object of the present invention is to improve a microstructure reactor of the generic type so that a greater catalyst surface is available in the reaction channels and negative influences caused by displacement of the catalyst material relative to the longitudinal axis of the reaction channel are avoided.

This object is achieved with a microstructure reactor of the generic type, in that the catalyst is not arranged in the reaction channel in the form of a metallic wire, but it is applied in a uniform layer thickness on the inside surface of the reaction channel. The electric heating of the silicon microstructure reactor is retained, in principle, so that no heat exchange through the reactor wall is required in order to carry out the endothermic reaction. Since there is no central catalyst wire in the reaction channels, there is no longer the related problem of its shifting from the center of the reaction channels due to thermal expansion. At the same time, this produces the great advantage that the effective surface of the catalyst is several times that of the known solution since, in accordance with this invention, the total inside surface of each reaction channel is coated with the catalyst.

It is essential for the operation and reliability of the reactors of this invention that the catalyst be applied in a uniform layer thickness. Due to the resulting local differences in electrical

resistance, a nonuniform layer thickness causes correspondingly different surface temperatures in the catalyst. A coating that is too thick results in a correspondingly low temperature, while a coating that is too thin can result in an elevated temperature that, in some cases, can even result in impermissible sintering of the catalyst. If the catalyst coating is completely absent in some spot, then there is also no electric heating there. At the same time, however, a correspondingly higher temperature will occur in the coated part at the same location in the circumferential direction along the length of the channel.

The catalyst is advantageously applied to a support material that adheres directly to the inside surface of the reaction channels as an intermediate layer. Such an intermediate layer preferably consists of Al_2O_3 or SiO_2 or mixtures of these two compounds, which are frequently utilized as catalyst supports in chemical units and are electrical insulators. For carrying the electrical heating current, however, it is also possible and frequently advantageous to use an electrically conductive support material. So that as uniform a temperature distribution as possible will be achieved, this support material should be applied in a uniform layer thickness, as in the case of the catalyst material. With regard to the resistance heating, it is clear that fluctuations in the layer thickness of the catalyst can practically be compensated by correspondingly inverse fluctuations in the layer thickness of the support material. The main idea is that the

sum of the two layer thicknesses at any given point should be as close as possible to that of the other locations. Care should be taken, however, to make sure the catalyst layer extends as continuously as possible over the entire inside surface of the reaction channels, in order to produce the optimum effect. To supply the electrical current for heating the reactor, electrical contacts are provided that are connected to the catalyst layer and/or to an electrically conductive intermediate layer. In order not to have a negative effect on the average cross section of the reaction channels, whose cross-sectional dimensions lie in the submillimeter range, it is a good idea to continue the catalyst layer and/or the layer of electrically conductive support material out of the reaction channels and to apply the electrical contacts outside the reaction channels per se.

For example, in the production of hydrogen by steam reforming of hydrocarbons, possible catalysts include, in particular, the metals in group VIII of the periodic table and their alloys with one another. Particularly preferable are the metals nickel and platinum and their alloys.

Building microvalves into the reaction channels or at least a part of the reaction channels makes it possible to control and regulate the flow of the process medium, which preferably consists of gases and/or vapors. Thus, by influencing the flow of the medium, there is a way to control the reaction and, indirectly, the temperature in the reactor. Of course, this can also be influenced by

proper control of the electrical current used for resistance heating of the catalyst layer and/or the electrically conductive intermediate layer. Another aspect is also of importance to the effectiveness of the reactor of this invention that is likewise aimed at influencing the current flow of the process medium. Producing microstructures on the inside surface of the reaction channels also provides turbulent flow on the part of the medium flowing through. Due to the constant mixing of the process medium this causes, the desired chemical reaction runs by and large to completion, so that a very high degree of conversion of the material used as the process medium can be achieved.

The reaction channels of the microstructure reactor are expediently produced parallel to one another in small plates of a silicon substrate by etching in a conventional manner. It is advisable to place the reaction channels upwardly open in the unmounted state on both the top and bottom of such a silicon plate. I order to produce packing of the reaction channels that is as dense as possible when the plate is thin, the reaction channels should be arranged such that immediately adjacent reaction channels have their open side facing to different sides of the silicon substrates. In this case, a microstructure reactor can easily be produced from a number of plates stacked one on the other which, in this way, are made so that the reaction channels that are open at the top and at the bottom are each closed off in the longitudinal direction by the "floor" of the plate

on top and the "cover surface" of the plate below and, thus, have an inlet and outlet opening for the process medium and reaction products only at the face ends.

In principle, the common methods of catalyst production can be used for applying an intermediate layer and catalyst layer. An electrically conductive intermediate layer can be applied by vapor deposition of metals and/or by electrodeposition methods. If the intermediate layer is to be made of aluminum oxide, the PVD method is expediently used. If the intermediate layer is made of metal, then electroplating is preferable.

The invention will be explained in greater detail below with the help of the exemplary embodiments in the drawing. It shows:

Figure 1: a schematic longitudinal section of the reaction channel of a reactor of this invention and

Figure 2: a schematic view of a plate-like silicon substrate with alternatingly produced reaction channels.

The schematic longitudinal section through flow channel **2** of a microstructure reactor made in accordance with this invention shown in Fig. 1 reveals various layers of the reactor wall. The first is the layer of silicon substrate **3**, on which catalyst **1** is applied. However, catalyst **1** does not adhere directly on the inside surface of silicon substrate **3**, but rather on a support material **4**, which is made, for example, of Al_2O_3 and, thus, is an electric insulator. Catalyst **1** is distributed with a uniform layer thickness over the inside surface of

reaction channel **2**. In order to introduce an electric current for heating the endothermic reactor, electric contacts **5** are provide, which are directly connected to the layer of catalyst **1**. If an electrically conductive material is used for support material **4** of catalyst **1**, electrical contacts **5** are also connected directly to support material **4**. If the layer of catalyst **1** and/or the layer of electrically conductive support material **4** are continued through the end faces of reaction channel **2** to the outside, then contacts **5** are applied outside the passage of reaction channel **2** and, as a result, they have no negative effect on the free passage surface for the process medium. The layer thickness of catalyst **1** and/or of an electrically conductive intermediate layer of support material **4** for catalyst **1** provide a parameter for influencing the temperature on the inside surface of the reactor of this invention. The thicker this layer or layers are, the lower the electrical resistance and, thus, the temperature for a given voltage will be.

Figure 2 shows a plurality of parallel reaction channels **2**, each of which has been produced in a plate **6** of silicon substrate, using the precision manufacturing methods known in silicon technology. Flow channels **2**, each of which has an open side **7** on one long side, are parallel and arranged alternatingly to one another in the sense that open side **7** of one reaction channels **2** always faces in the direction opposite that of the two immediately adjacent reaction channels **2**. In this way, reaction channels **2** are very densely packed. When a

plurality of such plates **6** are stacked one on the other, thereby producing a microstructure reactor in accordance with this invention, open sides **7** of flow channels **2** are each covered by a corresponding bottom of plate **6** above or below, resulting in a plurality of reaction channels **2** that are closed all around in the longitudinal direction. Only the two end faces are left open for the process medium to enter and for the reaction products to exit. Such a stack of plates can be packed, for example, in a metal housing that is provided with the required inlet and outlet for the medium supply. The cross-sectional dimensions of the flow channel lie in the submillimeter range and their length is preferably in the centimeter range. By connecting a plurality of such microstructure reactors in parallel, overall structures of any given size can be produced for greater throughput. It should be noted that, considering its size and weight, the reactor of this invention has an extremely high throughput capacity, particularly since the active catalytic surface is very large, compared to the reactor volume. Such a reactor is particularly suitable for mobile applications, such as for producing hydrogen for fuel cells in a vehicle.

Particularly good results are achieved if the inside surface of reaction channel **2** is provided with microstructures that cause turbulent flow in the medium stream.

There are several possibilities with regard to operating a microstructure reactor of this invention. In general, all the energy

required for carrying out the endothermic reaction of the process medium that is used is expediently supplied by the electric resistance heating. In many cases, however, it may also be advantageous to produce only a part of this energy by electrical means and to supply the remainder of the required heat, for example, by partial oxidation of the process medium. When the latter is expedient, the electrical heating can be limited only to an initial heating-up phase and for initiating the partial oxidation. Partial oxidation can be easily controlled, for example, by adding a measured amount of oxygen to a stream of hydrocarbons from which the hydrogen is to be produced, so that the oxygen can oxidize only a part of the process medium, thereby releasing a calculable quantity of heat energy.

Claims

1. A microstructure reactor for carrying out endothermic chemical reactions in the presence of a catalyst (1) that is electrically heatable by resistance heating, whereby the reaction chamber of the reactor comprises a plurality of reaction channels (2) that are produced in a silicon substrate (3) and that have cross-sectional dimensions in the submillimeter range, characterized in that catalyst (1) is applied in a uniform layer thickness to the inside surface of reaction channels (2).

2. A reactor as recited in Claim 1, characterized in that catalyst (1) is applied to a support material (4), which adheres in

the form of an intermediate layer directly on the inside surface of reaction channels (2).

3. A reactor as recited in Claim 2, characterized in that support material (4) is an electrical insulator, in particular Al_2O_3 and/or SiO_2,.

4. A reactor as recited in Claim 2, characterized in that support material (4) is electrically conductive and is applied in a uniform layer thickness.

5. A reactor as recited in one of the Claims 1 through 4, characterized in that the layer of catalyst (1) and/or the layer of electrically conductive support materials (4) are continued outwardly out of reaction channels (2) and that electrical contacts (5) for applying the heating current are attached outside reaction channels (2).

6. A reactor as recited in one of the Claims 1 through 5, characterized in that catalyst (1) is made of platinum and/or nickel.

7. A reactor as recited in one of the Claims 1 through 6, characterized in that at least one part of reaction channels (2) is provided with microvalves for controlling the medium flowing through the reactor.

8. A reactor as recited in one of the Claims 1 through 7, characterized in that the inside surfaces of reaction channels (2) are provided with microstructures for producing turbulent flow.

9. A reactor as recited in one of the Claims 1 through 8, characterized in that reaction channels (2) are made parallel to one another in a plate (6) of silicon substrate (3), that immediately adjacent reaction channels (2) are open alternatingly to the top and bottom side of plate (6), and that the reactor comprises a plurality of plates (6) of silicon substrate (3) stacked one on the other, whereby longitudinal openings (7) of reaction channels (2) are each covered by plate (6) located above and below.

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1 page of figures attached.

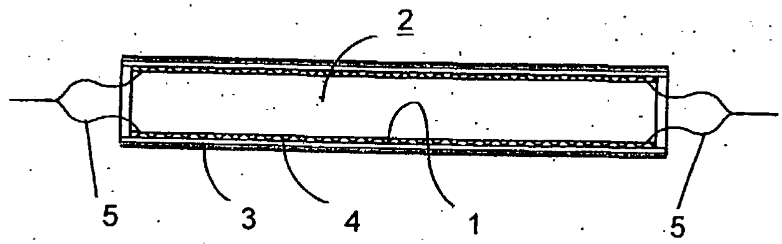

Fig. 1

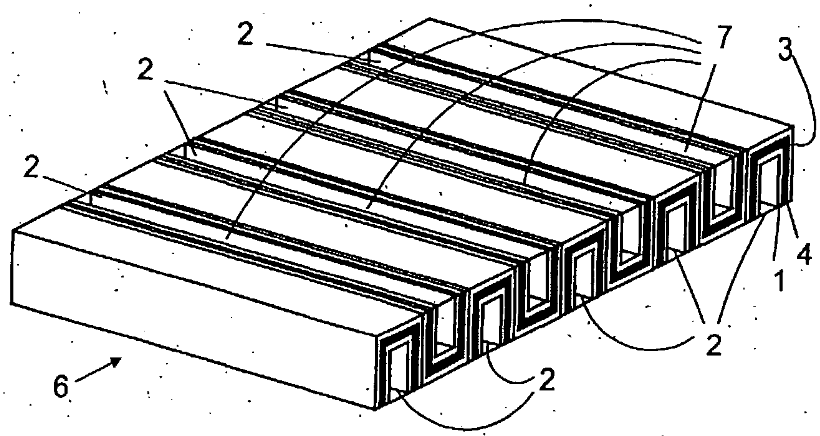

Fig. 2